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# InGaN/GaN ultraviolet LED with a graphene/AZO transparent current spreading layer

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**Abstract:** We report an approach of using an interlayer of single layer graphene (SLG) for electroluminescence (EL) enhancement of an InGaN/GaN-based near-ultraviolet (NUV) light-emitting diode (LED) with an aluminum-doped zinc oxide (AZO)-based current spreading layer (CSL). AZO-based CSLs with and without a SLG interlayer were fabricated on the NUV LED epi-wafers. The current-voltage (I-V) characteristic and the EL intensity were measured and compared. We find that the LED without the SLG interlayer can possess a 40% larger series resistance. Furthermore, a 95% EL enhancement was achieved by the employment of the SLG interlayer.

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**OCIS codes:** (230.0230) Optical devices; (230.3670) Light-emitting diodes.

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## 1. Introduction

Near-ultraviolet (NUV) light-emitting diodes (LEDs) have attracted significant research interest due to their applications in various fields including white LED light sources, detection systems of biochemical agents, non-line-of-sight communication, water purification and so on [1–3]. However, their performances are still limited due to the challenge of finding a highly conductive current spreading layer (CSL) to the NUV-LED with high NUV transparency. This issue comes from the difficulty in growing a highly Mg-doped p-GaN because of its high activation energy and the formation of Mg-H complexes [4, 5]. In addition, for GaN-based LEDs, it is difficult to find an appropriate CSL material having a sufficiently high work function for p-GaN, thus leading to a large Schottky barrier height (SBH) at the p-GaN/CSL interface [5–8]. The conventional Ni/Au CSL has a good electrical performance on p-GaN, but the low transparency in the NUV range hinders its use in NUV LEDs [8–10]. Today, indium tin oxide (ITO) with superior conductivity and transparency has been widely used as a CSL material in NUV LEDs [11–13]. However, the cost of ITO can grow high in the future due to the scarceness of indium while its thermal stability is not satisfactory [14, 15]. Aluminum-doped zinc oxide (AZO) is an alternative indium-free material, which has similar electrical and optical properties. It is also low-cost, nontoxic and more stable at high temperatures, that offers substantial attractions in NUV LEDs [16, 17]. Electrical characteristics could also be significantly improved by insertion of a Ni-based interlayer between the AZO film and the p-GaN layer [18–20]. This is due to the formation of Ga vacancies near the surface of p-GaN leading to a decreased contact resistivity. Here, we propose an approach to further improve the performance of AZO-based CSLs in NUV LED applications.

Single layer Graphene (SLG) is a two-dimensional carbon material consisting of a hexagonal array of carbon atoms, which is known for possessing outstanding properties including high carrier mobility, good thermal conductivity and mechanical stability [21–23]. Moreover, the high transparency in a wide spectral range including NUV makes it a promising transparent CSL material in NUV LED applications [24, 25]. Furthermore, in terms of the work function, graphene is more superior when compared to the reported work function of AZO [26–30]. This indicates a potential of being an effective interlayer to improve the performance of AZO by modifying the SBH. The SBH depends on the work function of the CSL material in contact and the sum of the electron affinity (4.1 eV) and the bandgap (3.4 eV) of the p-GaN (7.5 eV in total) [31–34]. In order to maximize the drive current and minimize the leakage current under a certain voltage, the work function of the CSL material is desired to be greater than 7.5 eV for p-GaN. Due to the difficulties of finding a conductive material with a work function larger than 7.5 eV, another option is to reduce the SBH by decreasing the difference through the employment of a material with a sufficiently high work function [31–34]. The reduction of the SBH can result in a lower contact resistance

and it has been reported that the contact resistance at the p-GaN/CSL interface decreases exponentially with increased work functions of CSL materials [5–8].

There have been works focused on combining graphene and AZO nanorods for improvement of LED device performance [35–37]. However, few research results have been reported on the combination of SLG and AZO as plain CSLs in NUV LED applications. In this work, we have investigated the effect of including SLG with AZO in a CSL for a NUV LED. AZO-based CSLs with and without a SLG interlayer were fabricated on both sapphire samples and NUV epi-wafers. After the fabrication, optical properties of the CSLs on sapphire samples and electrical properties on NUV epi-wafers were measured and compared.

## 2. Experimental details

### 2.1 Transferring graphene sheets to substrates

Pieces of 2'' × 4'' SLG grown by chemical vapour deposition (CVD) on Cu foil (GRAPHENE SUPERMARKET, Reading, Massachusetts, USA) was employed in the experiment. The SLG sheet was transferred to an NUV epi-wafer or a sapphire sample by a standard transfer process: at the beginning, a layer of 2  $\mu\text{m}$  photoresist AZ5214E was spin-coated onto the SLG and the AZ5214E/SLG stack was then released by etching the underlying Cu foil in a  $\text{Fe}(\text{NO}_3)_3$  solution (17 wt%) at room temperature. The AZ5214E/SLG stack floating in the solution was then washed by deionized (DI) water and transferred to the target substrate followed by drying at room temperature for 24 hours. Finally, the AZ5214E on the SLG was dissolved in acetone at room temperature.

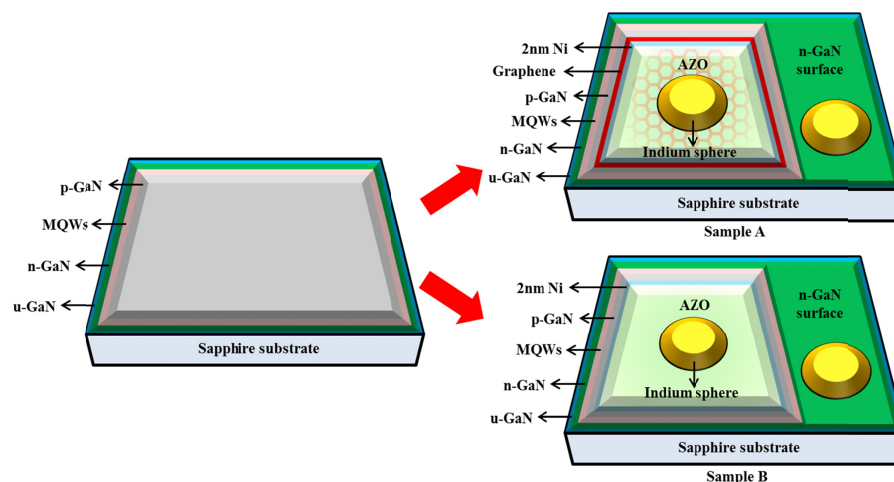


Fig. 1. Schematic illustration of the structure of the NUV epi-wafer (left) and the fabricated sample A with the CSL A of SLG/Ni/AZO and the sample B with the CSL B of Ni/AZO (right) both using indium spheres for carrier injection.

### 2.2 Fabrication of the current spreading layers

Two types of CSLs, which are SLG/2 nm Ni/250 nm AZO (CSL A) and 2 nm Ni/250 nm AZO (CSL B), were fabricated on both NUV epi-wafers and sapphire samples for measurements of electrical properties and transmittance, respectively. The InGaN/GaN NUV LED epi-wafers were grown on a c-plane (0 0 0 1) sapphire by metal-organic CVD CRUIS I (AIXTRON, Herzogenrath, Germany). The grown LED epi-layer consists of a sequence of a 3.5  $\mu\text{m}$  thick GaN buffer layer, a 2  $\mu\text{m}$  thick Si-doped n-GaN layer, nine periods of InGaN/GaN multiple quantum wells (MQWs), and finally a 130 nm thick Mg-doped p-GaN layer. In the fabrication process, first, a layer of 2 nm Ni was deposited by electron beam evaporation using Physimexa  $\Phi$ SES250 (Physimexa Technologie, Villiers le Bacle, France)



on the top of a NUV epi-wafer with a transferred  $6 \times 6 \text{ mm}^2$  SLG sheet. Afterwards, the Ni layer was treated by rapid thermal annealing using Jipelec JETFIRST (SEMCO Technologies, Montpellier, France) in air at  $525^\circ\text{C}$  for 5 minutes to increase its transparency. Subsequently, a layer of 250 nm AZO with a sheet resistance of  $70 \Omega/\text{sq}$  was deposited by a sputtering cathode TORUS (Kurt Lesker, Clairton, USA), using a ZnO target containing 2%  $\text{Al}_2\text{O}_3$  [38]. Identical fabrication steps were also applied on the NUV epi-wafer without the SLG and also on sapphire samples with and without a SLG. Consequently, two types of CSLs were fabricated on both the NUV epi-wafers and the sapphire samples. In the end, for the NUV epi-wafer with the CSL A (sample A) and for the NUV epi-wafer with the CSL B (sample B), a diamond pen was used to expose the n-GaN layer and indium spheres were added to their p-GaN and n-GaN surfaces for current injection, respectively, as shown in Fig. 1. In addition, silicon (Si) samples with the surface partially covered by gold (Au) and partially covered by SLG, AZO or Ni were fabricated assisted with standard photolithography and lift-off processes for work function measurements.

### 2.3 Characterization

The transmittance of the CSLs on the sapphire substrates was measured using an OL 700-71 6-inch diameter integrating sphere system (Gooch & Housego, Ilminster, UK) assisted with a Xenon lamp and a CAS 140 B optical spectrometer (Instrument Systems, Munich, Germany). The thickness of the transferred SLG sheet was characterized by Raman spectroscopy, using a DXRxi Raman imaging microscope (Thermo Scientific, Waltham, Massachusetts, USA). The Raman spectrum of the graphene was recorded with an integration time of 25 seconds, using a 633 nm laser with a power of 8 mW. The electroluminescence (EL) spectra were obtained using a fiber-coupled optical spectrometer. The current-voltage (I-V) data from the LEDs were obtained using a Model 2450 Interactive SourceMeter instrument system (Keithley, Solon, Ohio, USA). The work function measurements were carried out using PeakForce Kelvin probe force microscopy of a Dimension Icon atomic force microscope (AFM) (Bruker, Billerica, Massachusetts, USA).

### 3. Results and discussion

The transmittance for the two types of CSLs deposited on sapphire samples was measured in the wavelength range of 380-430 nm, as shown in Fig. 2. For the CSL B on sapphire, the transmittance is 66% at 386 nm while the CSL A on sapphire only suffers a small transmittance loss at 386 nm by adding the SLG interlayer and confirming the high transparency of SLG in the NUV range.

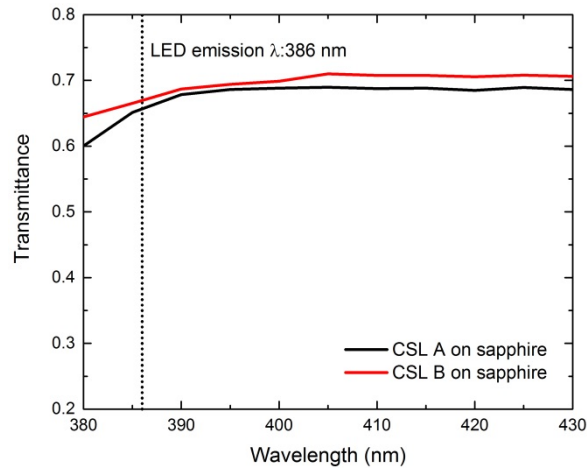


Fig. 2. Optical transmittance spectra of a SLG/Ni/AZO CSL (CSL A) and a Ni/AZO CSL (CLS B) on sapphire samples in a wavelength range of 380-430 nm.

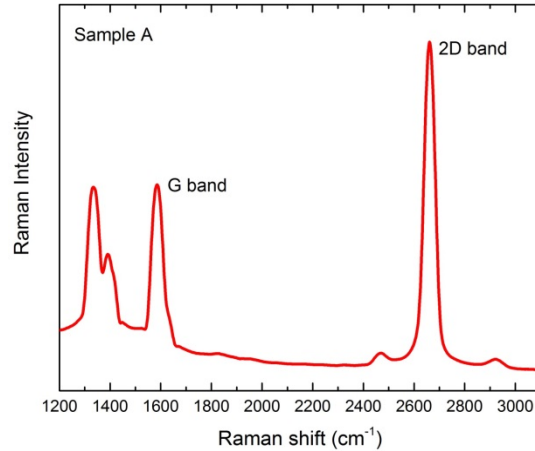


Fig. 3. Raman spectrum of the transferred SLG on sample A collected using a 633 nm laser with a power of 8 mW.

Figure 3 shows the Raman spectrum obtained by measuring the SLG transferred onto sample A. There are two dominant peaks which are the G peak at  $\sim 1580 \text{ cm}^{-1}$  and the 2D peak at  $\sim 2700 \text{ cm}^{-1}$  in the Raman spectrum of the SLG confirming the existence of the transferred SLG. The G to 2D peak intensity ratio identifies the thickness of the graphene layer. In our case, the value of  $I_G/I_{2D}$  is smaller than one ( $I_G/I_{2D} = 0.67$ ) and 2D-band has a full width at half maximum of  $\sim 60 \text{ cm}^{-1}$  indicating the graphene layer is a SLG [39, 40].

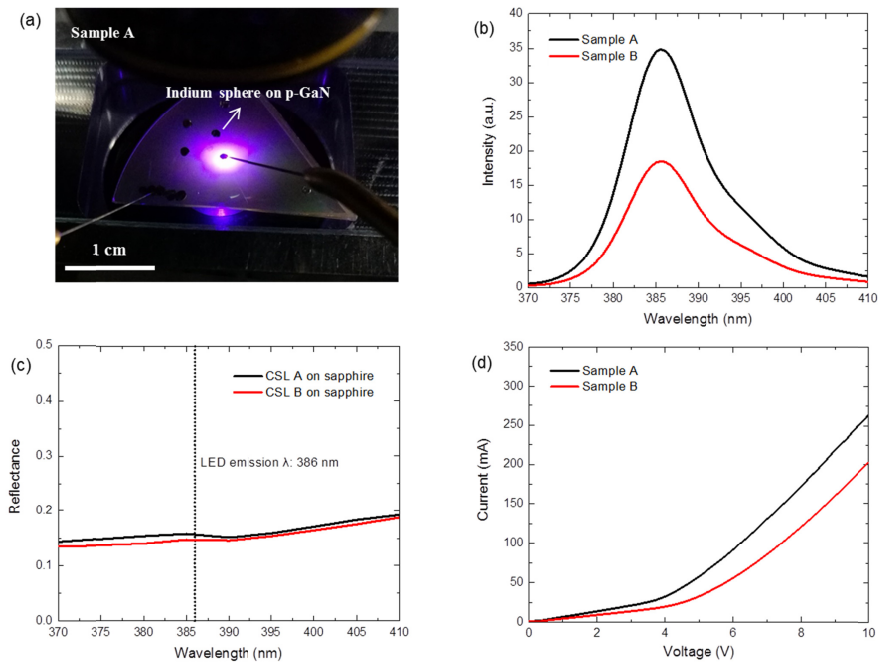


Fig. 4. (a) Photograph of sample A during light emission at an injection current of 50 mA; (b) EL spectra of sample A and B at an injection current of 50 mA; (c) Optical reflectance spectra of a SLG/Ni/AZO CSL (CSL A) and a Ni/AZO CSL (CLS B) on sapphire samples in a wavelength range of 370-410 nm. (d) I-V characterization of sample A and B for an input voltage range of 0-10 V.

The EL NUV emissions were obtained for both sample A and sample B. During the measurement, probes were pressed against the indium spheres on the p-GaN and the n-GaN layers for carrier injection. Figure 4(a) shows a photograph of sample A during light emission at an injection current of 50 mA. Figure 4(b) shows the EL spectra of sample A and sample B. The EL spectra were collected from the backside of the samples at a drive current of 50 mA and the peak emission was measured to be at around 386 nm. The emission intensity of sample A is 95% stronger than that of sample B at the peak wavelength. In addition, Fig. 4(c) shows a 0.5% reflectance difference at 386 nm between CSL A (15.5%) and CSL B (15%) on sapphire samples and this indicates that the 95% EL enhancement is not dominantly induced by the reflectance difference. Furthermore, the I-V characteristics of sample A and sample B were measured and the results are shown in Fig. 4(d). The current was measured by applying a voltage range of 0-10 V and the NUV emission starts at around 4 V for both samples. The forward voltage at an injection current of 50 mA was 4.6 V for sample A and 5.8 V for sample B. By fitting to the I-V curves after the turn-on, around 40% larger series resistance of sample B (37  $\Omega$ ) than that of sample A (26  $\Omega$ ) is shown due to the absence of a SLG interlayer in sample B.



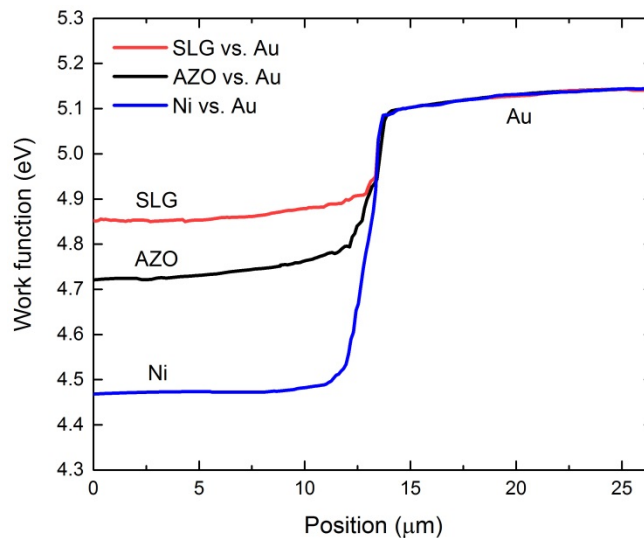


Fig. 5. Work functions of SLG, AZO and Ni (left region) in comparison with that of Au (right region).

To study the physics behind the EL enhancement by applying the SLG interlayer, work function measurements were carried out and the results are shown in Fig. 5. As indicated in the graph, SLG (4.85 eV) has a higher work function than Ni (4.48 eV) and AZO (4.74 eV). The Ni layer deposited for work function measurement was also treated by rapid thermal annealing in air at 525 °C for 5 minutes. The higher work function of SLG than that of Ni or AZO causes a reduction of the SBH at the interface of the contacting layer and the p-GaN consequently allowing an easier carrier injection process through the p-GaN layer [5–8]. A simplified performance comparison as a contact layer on p-GaN between SLG and AZO can be made. According to the reported curve in [6] demonstrating the relationship between work functions and contact resistances, the work function difference of 0.11 eV between AZO and SLG leads to a 1.5 times larger contact resistance of AZO on p-GaN. This can be estimated that, in contrast with AZO, the current through the SLG interlayer can be increased by 50% under an identical voltage when the other relevant resistances are kept identical. This estimated result is comparable to the 40% increase for the current measured on sample A at 8 V shown in Fig. 4(d). The comparison was made to AZO instead of Ni because in this work the employed 2 nm thin thickness and the 525 °C annealing temperature for Ni can lead to self-organization of Ni into nanoscale islands hence letting AZO in contact with p-GaN [41, 42].

#### 4. Summary

In summary, two types of CSLs which are SLG/Ni/AZO and Ni/AZO were successfully fabricated. This was done by using a standard graphene transfer process followed by deposition of Ni and AZO on both the p-GaN layer of the InGaN/GaN-based NUV-LED epi-wafers and sapphire substrates. The transmittance of the CSLs was measured and SLG shows a low transmittance reduction at a wavelength of 386 nm indicating its high transparency in NUV range. In addition, the graphene sheet was identified by micro-Raman spectroscopy confirming its type of SLG. In I-V characterization, it is shown that the LED without the SLG interlayer can possess a 40% larger series resistance. Furthermore, a 95% EL enhancement was achieved for the epi-wafer with the SLG interlayer. The improvement of EL and I-V performance can be explained by the high work function of SLG. Based on the optical and electrical characterizations, we conclude that SLG interlayers can improve the performance of NUV LEDs with AZO-based CSLs.

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